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Scattering of positrons by hydrocarbons at intermediate energies

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Abstract : Scattering of positrons by several hydrocarbons is investigated within the framework of Independent Atom Model (IAM) and the total cross sections (elastic + inelastic) have been obtained over a wide incident energy range (100–1000 eV). The present values of total cross section agree fairly well with the experimental data and other theoretical calculations available

Keywords : Positrons, hydrocarbons, independent atom model.

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1. Introduction

The $e^{\pm}$-molecule scattering in the intermediate energy range presents a more complex problem than the corresponding $e^{\pm}$-atom scattering due to its multicentre nature, the lack of a centre of symmetry and its nuclear motion. Hence for the above scattering, different approximate approaches have been proposed and developed. A simple method to investigate $e^{\pm}$-molecule scattering at intermediate and high incident energies, is the independent atom model [1]. Recently, Raj [2–4] has employed this approach with partial waves to calculate the total scattering cross sections for positron scattering by a number of molecules namely O_2, CO, CO_2, SO_2, CS_2, OCS, SF_6, H_2 and H_2O and for e^--scattering by a number of hydrocarbons at intermediate energies. The calculated values of the total cross section were found very encouraging and compared very well with the other theoretical calculations and the experimental data.

Recently, hydrocarbon molecules have received much interest not only as prototypes of polyatomic molecules but also as important species of planetary atmosphere and interstellar media. They play a very significant role in the chemistry of these environments [5]. For example, acetylene (C_2H_2) is a trace constituent formed by the photodissociation

of methane (CH_4) in the atmosphere of Jupiter, Saturn and Titan. Its concentration in the earth's atmosphere is expected to nearly double by the year 2030 due to increased use of automobiles. Hydrocarbons are also used in chemical vapour deposition (microchip) industry [6] where carbon produced in the low temperature discharge is used to harden the substrates. Reliable scattering cross sections of hydrocarbons are needed in fusion plasma devices where these are produced by the reaction of graphite with deuterium fuel [7,8]. So, it is worthwhile to extend this approach *i.e.* independent atom model to the calculation of the total cross sections for positron scattering by some of the hydrocarbons, namely, CH_4, C_2H_6, C_3H_8, C_4H_{10}, C_2H_4, C_3H_6, C_4H_8, C_2H_2 and C_6H_6 over a wide incident energy range (100–1000 eV). In this energy range, the only theoretical investigations available are of Baluja and Jain [9,10]. In an investigation [9], they have solved the scattering equation for positron-CH_4, C_2H_2 molecules in partial waves using variable phase approach to obtain the positron total cross section and in an another investigation [10], they have used an analytic formula to calculate positron-C_2H_4, C_2H_6 and C_3H_6 cross sections. On the other hand, we have solved the problem for positron-constituent atoms of the molecule and employed independent scattering centre approximation to obtain molecular total cross section.

2. Theory

The elastic scattering amplitude $F_m^{el}(\theta)$ of a molecule with N atoms in independent atom model (IAM) whose nuclei are assumed to be fixed in space is given by [3]

$$F_m^{el}(\theta) = \sum_{j=1}^{N} f_j^{el}(\theta) e^{iK \cdot r_j}, \tag{1}$$

where $f_j^{el}(\theta)$ is the elastic scattering amplitude proper to the j-th atom of the molecule. K is the momentum transfer vector and r_j is the position vector of the j-th atom with respect to some arbitrary origin, preferably the centre of mass of the molecule. It may be noted that the eq. (1) neglects the multiple scattering [11,12] and the valence bond distortion [13] effects. The likely effect of both is to reduce the value of cross section. However, their effect is supposed to be negligible at high incident energies. The total cross section $Q_T(E)$ (elastic + absorption) of the molecule using the optical theorem then can be obtained from the relation

$$Q_T(E) = 4\pi/k.\ \mathrm{Im}\, F_m^{el}(\theta = 0) = 4\pi/k. \sum_{j=1}^{N} \mathrm{Im}\, f_j^{el}(\theta = 0), \tag{2}$$

where k is the incident positron wave number.

The atomic elastic scattering amplitude $f_j^{el}(\theta)$ in partial waves is given by

$$f_j^{el}(\theta) = 1/k \sum_{l=0} (2l+1) \left(e^{i\delta_{lj}} \sin \delta_{lj} - \delta_{lj}^{B} \right) P_l(\cos\theta) + f_{dp}^{Bj}(\theta), \tag{3}$$

where $f_{dp}^{Bj}(\theta)$ and δ_{lj}^{B} are the first Born scattering amplitude and the phase shifts, respectively due to long range polarization potential only. The above equation takes the first

L_{max} partial waves phase shifts exactly whereas the contribution of the higher remaining ones is included through the first Born approximation along with the approximation that $e^{i\delta_l} \sin \delta_l \equiv \delta_l$. The value of L_{max} is so chosen that at $l = L_{max}$ the difference between the real part of δ_{lj} and $\delta_{lj}^B \leq 2$ percent. If this condition is not satisfied at any energy the maximum value of L_{max} is taken to be 30.

To obtain δ_{lj}, the phase shift for the l-th partial wave of the j-th atom of the molecule, the following radial equation

$$\left[d^2/dr^2 + k^2 - V_{opt}(r) - l(l+1)/r^2\right] U_l(r) = 0 \tag{4}$$

is solved numerically under the following boundary conditions

$$U_l(r) \underset{r\to 0}{=} r^{l+1}$$

and

$$U_l(r) \underset{r\to \alpha}{=} 1/k.\ \sin(kr - l\pi/2 + \delta_l), \tag{5}$$

where δ_l is the phase shift for the l-th partial wave. k^2 is the energy of the incident positron (we employ atomic units in which the length is expressed in a_o and the energy in rydberg). $V_{opt}(r)$ is the optical potential. It is complex, spherically symmetric and energy dependent and is given by

$$V_{opt}(r) = V_{st}^+(r) + V_{pol}^+(r) + iV_{abs}^+(r), \tag{6}$$

where V_{st}^+, V_{pol}^+ and V_{abs}^+ are the static, polarization and absorption potentials, respectively for $e^+ - j$-th atom. The polarization effects are included by taking spherically symmetric and energy dependent potential suggested by Jhanwar and Khare [14] :

$$V_{pol}^+(r) = -\alpha_d r^2/(r^2+d^2)^3 - \alpha_q r^4/(r^2+d^2)^5 \tag{7}$$

$$\text{with } d = 0.75k/\nabla,$$

where α_d and α_q are, respectively the dipole and quadrupole polarizabilities of the atom and ∇ represents the mean excitation energy of the atom which may be calculated from the ground state properties of the atom. Thus, it may be noted that the present polarization potential involves no free parameter. Since the polarization potential is a second order potential, and we have considered only dipole and quadrupole components of the polarization potential, it is expected that there will not be much difference between two polarization potentials, one for electron and other for positron.

As we know that the static potential for positron will have opposite sign to that for electron. The analytical expressions of Salvat *et al* [15] obtained for the static potential of electron by fitting procedure to the Dirac-Hartree-Fock-Slater (DHFS) self-consistent data, have been used in the present investigation. Thus, the static potential appropriate to positron scattering is given by

$$V_{st}^+(r) = 2Z/r \cdot \sum_{i=1}^{3} A_i e^{-\alpha_i r}. \tag{8}$$

The values of the parameters A_i and α_i are given by Salvat *et al* [15] and Z is the atomic number of the atom.

For absorption potential, we use the model suggested by Baluja and Jain [10]. They employed a local absorption potential derived semi-empirically from the electron absorption potential $V^-_{abs}(r)$ of Staszewska *et al* [16]. In a sequence of development of the corresponding absorption potential $V^+_{abs}(r)$ for positron, Baluja *et al* [17] first used the following form

$$V^+_{abs}(r) = 4/kr \cdot V^-_{abs}(r). \tag{9}$$

But with this form of absorption potential, the results were not found much encouraging. They suggested that actual numerical values of $V_{abs}(r)$ for electron and positron are different. Also, the actual relationship between $V^+_{abs}(r)$ and $V^-_{abs}(r)$ is not known. Therefore, Baluja and Jain [9,10] assumed the following form for $V^+_{abs}(r)$

$$V^+_{abs}(r) = 2/\sqrt{kr} \cdot V^-_{abs}(r). \tag{10}$$

This form of $V^+_{abs}(r)$ was found very successful in reproducing the experimental data for a large number of targets. Hence, the use of the factor $2/\sqrt{kr}$ is justified and the same form has been used in the present work also.

$V^-_{abs}(r)$ is a function of charge density, incident electron energy and the mean excitation energy ∇ of the target and given as

$$V^-_{abs}(r) = -\rho(r)T^{1/2}_{loc}\, 8\pi/5k^2k_f^3 \cdot H(x)[A1 + A2 + A3], \tag{11}$$

where $A_1 = 5k_f^3/\nabla$,

$$A_2 = -k_f^3(5k^2 - 3k_f^2)/(k^2 - k_f^2)^2,$$

$$A_3 = 2H(y)y^{5/2}/(k^2 - k_f^2)^2,$$

where $x = k^2 - k_f^2 - \nabla$ and $y = 2k_f^2 + \nabla - k^2$

and $\rho(r)$ is given as

$$\rho(r) = 2Z/4\pi r \cdot \sum_{i=1}^{3} A_i\alpha_i^2 e^{-\alpha_i r}. \tag{12}$$

The T_{loc} is the local kinetic energy of the incident positron and $H(x)$ is the Heaviside unit step function.

Finally having obtained $F^{el}_m(\theta)$, the total cross sections for positron scattering by first four alkanes, three alkenes, one alkyne and one arene were calculated using eq. (2). The present values of total cross section over a wide incident energy range (100–1000 eV) along with experimental and other theoretical values are shown in Figures (1–4).

3. Result and discussion

Figures (1–2) show the e^+-alkanes total cross sections along with several sets of experimental data and other theoretical results. The present values of total cross section for

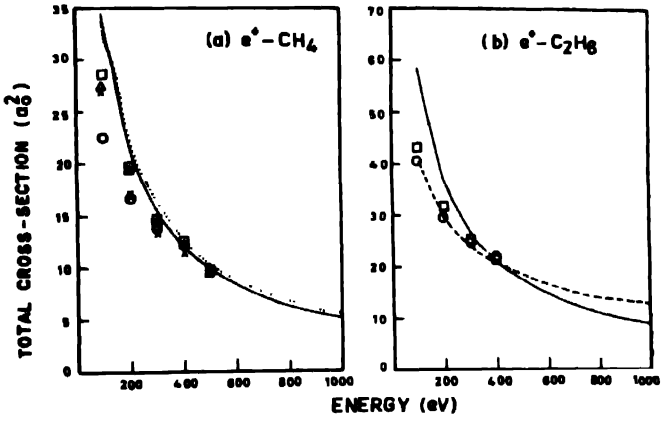

Figure 1. Total cross sections e^+-CH_4 and e^+-C_2H_6 at various impact energies

Theory —— Solid curve : Present work
Dotted curve . Ref. [9]
– – – – Broken curve Ref. [10]

Experimental data Δ, Ref [18]
□, Ref [19]
O, Ref [20]
X, Ref [21]

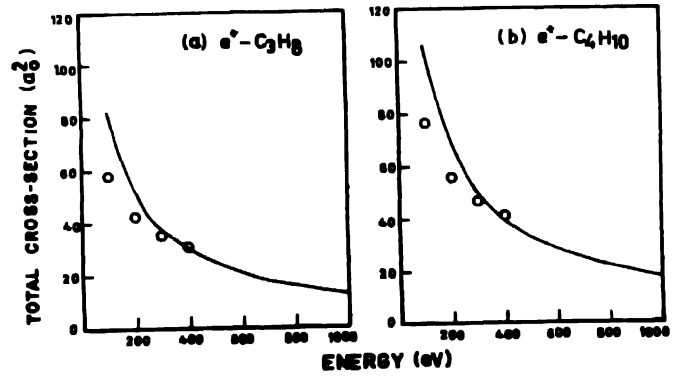

Figure 2. Total cross sections for e^+-C_3H_8 and e^+-C_4H_{10} at various impact energies. Same as above.

all the four alkanes, methane (CH_4), ethane (C_2H_6), propane (C_3H_8) and butane (C_4H_{10}) are in satisfactory agreement with the available experimental data [18–21]. Present values of total cross section for e^+-CH_4 scattering (Figure 1a) agree fairly well with all the available data except at 100 eV. Our values are more close to the data of Sueoka and Mori [19] than any other data. Furthermore, the present value of total cross section for positron-CH_4

scattering are in excellent agreement with the theoretical values of Baluja and Jain [9] throughout the whole energy range investigated. It may be noted that Baluja and Jain have used the molecular wavefunctions to generate positron-CH_4, C_2H_2 interactions and employed partial wave method to solve scattering equation. On the other hand, we use rather

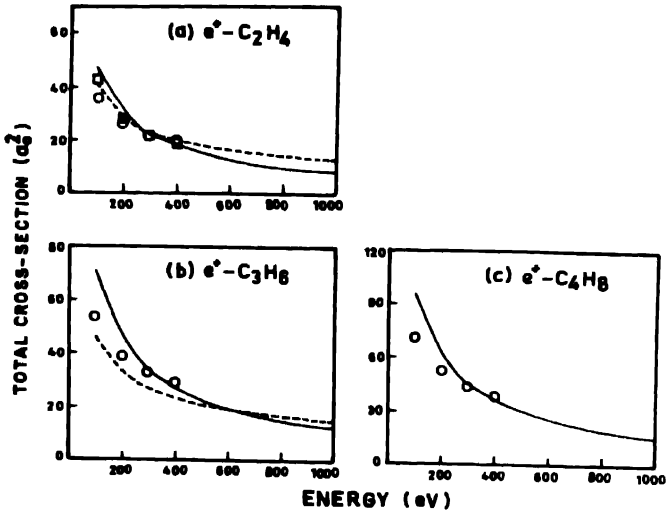

Figure 3. Total cross sections for e^+-alkenes at various impact energies
Same as above

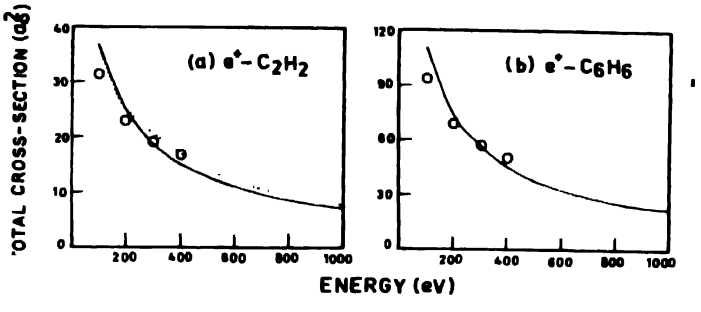

Figure 4. Total cross sections for e^+–C_2H_2 and e^+–C_6H_6 at various impact energies
Same as above.

simple approach (IAM) in which the problem is reduced to positron-atom scattering. In the present investigation, the scattering equation was solved again in partial waves but for the constituent atoms of the molecule and employed Independent Atom Model to obtain the molecular scattering amplitude. Figure 1b also shows the total cross section for positron-C_2H_6 scattering and the present results are in good agreement with both the experimental data available for C_2H_6 *i.e.* Sueoka and Mori [19] and Floeder *et al* [20]. Our values slightly overestimate the data at energies ≤ 200 eV. The theoretical values of Baluja and Jain [10] are also comparable with the present values for e^+-C_2H_6 for the incident energies

$E > 200$ eV. However for incident energies $E \leq 200$ eV, present values are slightly higher than the values of Baluja and Jain. In case of C_3H_8 and C_4H_{10} (Figure 2), no other theoretical work is known in the present energy range and only the data of Floeder *et al* [20] is available. The present results for these molecules show the similar type of agreement with the data as obtained in case of CH_4 and C_2H_6.

It may be noted that the general trend of agreement between the present values of total cross section for positron scattering by alkenes (C_2H_4, C_3H_6, C_4H_8) and the experimental data (Figure 3) is almost similar as obtained for alkanes. For ethene (C_2H_4) our values are again in better agreement with the data of Sueoka and Mori [19] than the data of Floeder *et al* [20]. Our values are in good agreement with the theoretical values of Baluja and Jain [10] also at all the incident energies investigated here (Figure 3a). For propene (C_3H_6), only the experimental data of Floeder *et al* is available. It is evident from Figure 3b that the present results are in better agreement with this data than the theoretical values of Baluja and Jain. It may be noted that Baluja and Jain [10] have obtained total cross section for these molecules (C_2H_6, C_2H_4, C_3H_6) using analytical formula (see eq. (2.3) of ref. [10]). For C_4H_8 molecule (Figure 3c), no other theoretical investigation seems to be available. Our values are in satisfactory agreement with the available data of Floeder *et al* [20] for incident energies $E \geq 200$ eV.

It may be noted that for acetylene (C_2H_2) and benzene (C_6H_6), no experimental data is available to our knowledge, in the present energy range. However just to have a comparison, we have extrapolated the data of Floeder *et al* [20] for these two molecules. It may be noted that Floeder *et al* tried several relations and obtained a best fit to their data (see eq. (2) of ref. [20]) which involves number of positrons of the molecule and three other constants. Furthermore, the proposed formula described all their e^+-total cross section between 100 and 400 eV reasonably well. Figure 4 shows that the present values of total cross section for C_2H_2 and C_6H_6 are in satisfactory agreement with the extrapolated data of Floeder *et al* like other hydrocarbons investigated here. Furthermore, the present values are in excellent agreement with the theoretical results of Baluja and Jain [9] for C_2H_2 at all the incident energies investigated. For benzene, no other theoretical investigation is available to our knowledge, in the present energy range; hence no further conclusion may be drawn.

It is concluded that in general, the present approach yields the values of total cross section for positron scattering by hydrocarbons in good agreement with the experimental data for the incident energies $E \geq 200$ eV, for all the hydrocarbons investigated. However, the agreement is less satisfactory for incident energies $E \leq 200$ eV. The reason may be the multiple scattering effects which were neglected in the present investigation, the possibility of which increases with decreasing energy. The likely effect of multiple scattering is to reduce the cross sections. It is evident from Figures 1–4 that as the number of scattering centres increases, the disagreement between the present results and data increases at lower

incident energies where multiple scattering is supposed to play an important role. The agreement between theory and experiment is expected to improve even below 200 eV on inclusion of these effects. Furthermore, it may also be noted that our approach which is rather simpler, yielded almost same values of total cross sections as obtained by Baluja and Jain [9,10] through a somewhat difficult approach using full molecular wavefunctions. A good agreement between the results obtained by additivity rule, IAM and full molecular wavefunctions shows that in the energy range under investigation, it is not essential to use molecular wavefunctions. Simple calculations using additivity rule and IAM give equally good result. Coincidentally for total cross section, additivity rule and IAM give same results. It shows that the interference effects between the scattered waves originating from different atoms of the molecule do not give significant contributions as far as the calculations of total cross section is concerned. Furthermore, at intermediate and high incident energies, the projectile will reach closer to the target and it will see the molecule as independent atoms. That is why the results of IAM are expected to agree with the full molecular wave function calculations. However at low incident energies, the projectile will remain far away from the target and will see it as a molecule. At such energies, the IAM results are not expected to agree with the full molecular wave function calculations. It may also be noted that the molecules with large geometrical cross sections also have large positron scattering cross sections.

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